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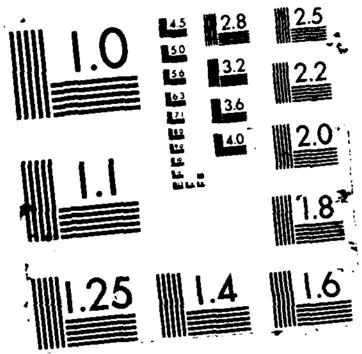
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Work is summarized in areas of:
1. Laser-Assisted Collisions
2. Velocity-Changing Collisions in Atomic Vapors
3. Coherent Transients
4. Collision-Induced Extra Resonances
5. "Breakdown" of the Optical Bloch Equations

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6. Saturation Spectroscopy

7. Dynamics of Two-and Three-level Atoms Interacting with Radiation Fields

8. Pulse Propagation. (Keywords) →

FINAL REPORT (FR)

Title: "Theoretical Studies Relating to the Interaction of Radiation with Matter: Atomic Collision Processes Occurring in the Presence of Radiation Fields"

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The Final Report for Contract No. N00014-77-C-0553 is divided into eight subject areas as follows:

1. Laser-Assisted Collisions
2. Velocity-Changing Collisions in Atomic Vapors
3. Coherent Transients
4. Collision-Induced Extra Resonances
5. "Breakdown" of the Optical Bloch Equations
6. Saturation Spectroscopy
7. Dynamics of Two-and Three Level Atoms Interacting with Radiation Fields
8. Pulse Propagation

A detailed description of the research carried out under this Contract can be found in the articles listed at the end of this report as well as in Annual Reports AR1-AR9 associated with the Contract. In this Final Report, the research areas are summarized briefly, with references to the appropriate articles. Advances achieved during the last year of the Contract are indicated where relevant, in each subject area.

1. Laser-Assisted Collisions

The theory of two types of Laser-Assisted Collisions has been developed. In Collisionally-Aided Radiative Excitation (CARE), collisional translational energy is used to provide the energy mismatch to allow radiation to be emitted or absorbed by an off-resonant radiation field. In Light-Induced Collisional Energy Transfer (LICET), two atoms collide in the presence of a radiation field and absorb or emit photons to change the states of both of the atoms - the reaction is energetically forbidden in the absence of either the collision or the radiation field.

A summary of our research in this field can be found in three review articles[27,46,78*]. A basic theory of CARE for physical interaction potentials was described[1], as well as theories involving approximate interaction potentials that allow for analytic solutions.[2,9] These solutions enabled us to predict the dependence of CARE and LICET cross sections on parameters such as laser field strength and laser-field-atom detuning. Our discussion of CARE was extended to "three-level" atoms and two incident laser fields, where new types of resonance behavior and interference effects were predicted[5,11].

For the LICET reaction, we were able to show that magnetic-state coherence could be generated in a laser-assisted collision[12,13], providing an additional important probe of the interatomic potential (similar methods have been applied subsequently to CARE under the general title of "half-collisions"). We are extending the calculation, carried out for the LICET line core to the line wings.[78] New physical phenomena appear which are absent in the analogous CARE calculations.

In the last year, we have modified the traditional LICET "two-state" calculation to include the effects of a non-virtual intermediate state.[73,74,78] Using a very simple model, we were able to resolve a long-standing discrepancy between theory and experiment on the fall-off of the LICET cross section with atom-field detuning.

Since atoms acquire or give up translational energy in a CARE reaction, we proposed CARE as a means for heating or cooling a vapor.[29,48] Heating

* Superscripts refer to the numbers in the list of publications at the end of this report.

of a vapor via CARE was demonstrated experimentally in the Na - noble gas collisions[26,31,32], but cooling was not observed (owing to dimmer formation). In any event, collisional cooling does not seem to give as effective a means for cooling as does the photon recoil technique.

2. Velocity-Changing Collisions in Atomic Vapors

A major emphasis of our research program was to understand the role that collision dynamics in an atomic vapor plays in modifying linear and non-linear spectroscopic line shapes. We were able to provide a physical picture that led to a clear understanding of the role of velocity-changing collisions on both level populations and electronic state dipole coherence. Using simple quantum-mechanical arguments[22], we explained that velocity-changing effects on level populations could be separated into classical and quantum-mechanical scattering regions, while the effects on dipole coherence led to a loss term corresponding to classical scattering and a diffractive term corresponding to quantum-mechanical scattering. Our results are summarized in a long review article[44] which provides a synthesis of the theory of velocity-changing collisions in atomic vapors and their influence on linear and non-linear spectroscopic line shapes.

The predictions of the theory for electric dipole coherence was verified in photon echo experiments involving Li-rare gas collisions.[18,19] Velocity-changing and fine-structure changing collisions involving atomic state populations in excited sodium were studies theoretically[6] and experimentally[7,8] using saturation spectroscopy. Recently, a more extensive study of velocity-changing collisions in ground and excited-state Na-rare gas collisions was carried out.[77] Excellent agreement between theory and experiment was achieved using model collision kernels with a minimum number of free parameters. An important parallel calculation[70] associated with this work led to new relationships between collision kernels and transport coefficients that could be used to reduce the number of free parameters in fitting data.

The role of velocity-changing collisions on magnetic-state coherence was also investigated.[20] This work stimulated several subsequent

experimental and theoretical investigations, but a simple picture of the manner in which velocity-changing collisions modify magnetic-state coherence is not yet available.

In the last year, we completed a calculation of the effects of velocity-changing collisions on electric-dipole coherence in strong fields. Specifically, we obtained expressions for the photon echo signal when the second excitation pulse in the excitation scheme is intense and of long duration. "Quenching" of the effects of velocity-changing collisions in strong fields are now understood. Articles relating to this work are in preparation.

It should be noted that our work on velocity-changing collisions should find renewed life in neutral particle traps. For "cold" atoms in such traps, scattering is quantum-mechanical in nature (owing to the large De Braglie wavelength), necessitating a quantum-mechanical picture of the role of velocity-changing collisions.

3. Coherent Transients

Several calculations were carried out to investigate new phenomena that could arise in coherenttransient spectroscopy. It was shown that photon echo experiments using standing-wave excitation fields would lead to multiple photon echoes that could be used to probe velocity-changing collisions.[3] Moreover, photon echoes in three-level atoms using double-resonance optical pulses[57] or one CW and one pulsed field[52] were studied. New echo phenomena were predicted which could be explained on simple physical grounds using Doppler phase diagrams and a dressed-atom picture of the atom-field interaction.

A theory of coherent transients for atoms prepared in pure dressed states of the atom plus radiation field has been developed. By controlling the initial conditions of the (atom + field), one can significantly modify the subsequent evolution of the system. The transient spectral response

of the system can differ dramatically from the steady-state results. Specifically, we studied the transient build-up of (a) the Autler-Townes doublet in the pump-probe spectroscopy of a three-level atom[61], (b) the resonance fluorescence triplet in a strongly-driven two-level atom[62] and (c) the probe absorption signal on a transition driven by a strong pump field.[76] Experimental verification of our predictions for the first[69] and second cases has been reported.

4. Collision-Induced Extra Resonances

In the presence of collisions, new structure can appear in various spectroscopic line shapes. An example of such a structure is the absorption resonance on the 3P - 4D transition in Na when an <u>off-resonant</u> field drives the 3S - 3P transition. An experimental investigation of this collision-induced resonance was carried out, giving results in excellent agreement with theory.[4] Related induced resonant structure was also predicted in four-wave mixing.[15]

Bloembergen and coworkers proposed and carried out a series of experiments involving extra resonances in four-wave mixing. These resonances were essentially collision-induced Rayleigh or Raman resonances, rather than the collision induced electric dipole resonances we had previously studied. We extended Bloembergen's work to cases where electronic state coherences could be produced in collisionally-aided reactions.[33] The macroscopic electronic-state coherences could then lead to coherent emission at optical frequencies. The magnetic degeneracy of the levels must be included in these calculations for a proper treatment of the problem.[37] (An irreducible tensor representation of the interatomic potential[34] is especially helpful in performing the calculation.) Experimental verification of our predictions was achieved on the 4D - 3P transition in Na, giving the first example of collision-induced coherent emission at an optical frequency.[41,42]

We are still working on an interpretation of some of these extra resonances. To further explore the collision-induced resonances, we

calculated the probe absorption on a transition simultaneously driven by a strong pump field. Collision-induced dispersive structure could be seen at the Rayleigh resonance frequency[45,46] with gain possible. The theory was carried out for an inhomogeneously broadened atomic sample[80], including effects of magnetic degeneracy.[79] It was shown that extra-resonances occur in the absence of collisions if the total magnetic-state alignment or orientation is not conserved.

Our work was extended to include the effects of velocity-changing collisions. We have seen that, in a two-level system whose total population is conserved, volicity-changing collisions lead to new resonance structures. An article on this subject is in preparation.

5. "Breakdown" of the Optical Bloch Equations

Renewed interest in the validity conditions for the optical Bloch equations followed an experiment by DeVoe and Brewer on free-induction decay in an impurity-ion crystal. We were able to show that the optical Bloch equations are valid in a limited domain only.[55,58,59] Whenever perturbations (velocity-changing collisions, magnetic fields) lead to frequency jumps in atomic transition frequencies, the Bloch equations go over into integral-differential equations. The manner in which strong radiation fields can "quench" the effects of these jump perturbations was explained using simple physical arguments relating to the relevant time scales in the problem.

In the weak-field regime, general expressions for the various propagators describing the effects of the jump perturbations were derived.[60] Strong field calculations using numerical methods are currently in progress.

6. Saturation Spectroscopy

Some general calculations relating to saturation spectroscopy were also carried out. A detailed description of saturation spectroscopy using a semi-classical dressed atom approach was developed and compared with the

"bare-atom" approach.[28] Both homogeneously and inhomogeneously broadened media were considered. This work has proved to be an invaluable reference for both CW and transient calculations. A theory of saturation spectroscopy in "three-level" atoms, including effects of magnetic-state degeneracy was developed.[35] The Autler-Townes doublet was found to split into four or six peaks as a function of the pump field's polarization - the splitting could be used to measure various oscillator strengths or laser field amplitudes. Analytic expressions for the line splittings were given for certain J values of the levels. Both irreducible tensor and standard (m-basis) representations were used. A review of optical pumping using lasers (including a discussion of collision-induced Rayleigh resonances) was given.[56] Finally an extensive calculation of pump-probe spectroscopy of a single transition, including effects of inhomogeneous broadening with arbitrary ratios of Doppler to natural width was carried out.[80] New structure in the probe absorption spectra is predicted as a result of this work.

7. Dynamics of Two-and Three-Level Atoms Interacting with Radiation Fields

The problem of a two-level atom interacting with a smooth off-resonant pulsed radiation field is a prototype for the general problem of two levels coupled by an interaction pulse. For this fundamental problem, it is surprising that the only pulse shape for which an analytic solution existed was a hyperbolic secant pulse, giving rise to the so-called Rosen-Zener solution. We were able to find an infinite class of smooth pulse functions for which analytic solutions could be obtained.[16] This work has stimulated a large number of articles for which analytic solutions could be obtained using various combinations of pulse and atom-field detuning envelope functions. We have extended our original work[21,28,36,39,42,47,49,51,63,64] and also obtained an analytic solution for a special three-level (or n-level) quantum system.[76]

8. Pulse Propagation

A calculation on pulse propagation in a medium of three-level atoms is still in progress.[68] We hope to clearly identify the interchange of energy between a "pump" and probe beam, including effects of diffraction, pump depletion, and dispersion.

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